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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Serial No.	:	09/415,781
Filed	:	October 8, 1999
Applicant	:	Zafar Iqbal, et al.
Title	:	Corrosion Resistant Coated Fuel Cell Plate With Graphite Protective Barrier and Methods of Making Same
TC/AU	:	1745
Examiner	:	T. Dove
Docket No.	:	19441-0019
Customer No.	:	29052

DECLARATION UNDER 37 C.F.R. § 1.131

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

We, Zafar Iqbal, James V. Guiheen, Dave Narasimhan, and Timothy Rehg, hereby declare that:

1. We are joint inventors of the above-referenced patent application. At the time of the invention, we were employed by AlliedSignal, Inc., which merged with and became known as Honeywell International, Inc. Subsequently, GE Power Systems acquired the business operations, including rights in this patent application, from Honeywell.

2. We have reviewed U.S. Patent No. 6,291,094 to Yoshimura et al. ("Yoshimura"), which was cited in the Office Action mailed June 9, 2003, in connection with the patent application. We understand that Yoshimura has an effective prior art date of May 3, 1999.

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conceived and reduced to practice the methods for corrosion passivation and manufacture of bipolar plates for a fuel cell defined by claims 1-25 prior to May 3, 1999. These documents are described below.

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PAGE 12/45 * RCVD AT 2/12/2004 5:48:50 PM [Eastern Standard Time] * SVR:USPTO-EFXRF-1/0 * DNIS:8729306 * CSID:4048538806 * DURATION (mm-ss):08-44

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9. I declare that all statements made herein of my own knowledge and belief are true and that all statements made on information and belief are believed to be true, and further that the statements are made with the knowledge that willful false statements are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such

willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Zafar Iqbal
ZAFAR IQBAL

8 September, 2003
Date

JAMES V. GUIHEEN

Date

DAVE NARASIMHAN

Date

TIMOTHY REHG

Date

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ZAFAR IQBAL	Date
James V. Guiheen JAMES V. GUIHEEN	9/8/03 Date
DAVE NARASIMHAN	Date
TIMOTHY REHG	Date

TOTAL P.05
PAGE 18/45 * RCVD AT 2/12/2004 5:48:50 PM [Eastern Standard Time] * SVR:USPTO-EFXRF-1/0 * DNIS:8729306 * CSID:4048538806 * DURATION (mm-ss):08-44

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ZAFAR IQBAL	Date
JAMES V. GUIHEEN	Date
Dave Narasimhan DAVE NARASIMHAN	9/3/03 Date
TIMOTHY REHG	Date

AO 983935.1

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U.S.S.N. 09/415,781
Filed: October 8, 1999
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JAMES V. GUIHEEN	Date
DAVE NARASIMHAN	Date
Timothy Rehg TIMOTHY REHG	Sept. 8, 2003 Date

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EXHIBIT A

AlliedSignal

Invention Disclosure

I.D. No. [to be supplied by Patent Dept.]

1. **Date:** [redacted]

2. **Title of Invention:**
   An Aluminum-Based PEM Fuel Cell Bipolar Plate

3. **Inventor(s):**

   a) **Name:** Zafar Iqbal **Phone/Fax:**(973)455 3899

   Address: 18 Erskine Dr
   Morristown NJ 07960 **S.S.#:** [redacted]
   County: Morris **Supervisor:** Mike Shand

   Citizenship: USA

   b) **Name:** Jim V. Guiheen **Phone/Fax:**(973)455 3935

   Address: 13 Rosemont Ave **S.S.#** [redacted]
   Madison NJ 07940
   County: Morris **Supervisor:** Santosh Das

   Citizenship: USA

   c) **Name:** Dave Narasimham **Phone/Fax:** (973)455 5171

   Address: 6 Summit Trail
   Flemington NJ 08822 **S.S.#** [redacted]

   County: Hunterdon **Supervisor:** Santosh Das

   Citizenship: USA

   d) **Name:** Tim Rehg **Phone/Fax:** (310) 512 2281

   Address: 26756 Basswood Ave
   Rancho Palos Verdes CA 90275 **S.S.#** [redacted]

   County: Los Angeles **Supervisor:** Nguyen Minh

Citizenship: USA

4. **Originating SBU:** Aerospace Equipment Systems

5. **Briefly describe the technical or commercial problem or need that this invention is intended to solve:** Bipolar plates that are corrosion-resistant and have sufficient electrical conductivity to provide high performance in a proton exchange membrane (PEM) fuel cell are composed of graphite. Graphite plates are heavy, bulky and expensive to manufacture. Noble metals like platinum are highly corrosion-resistant and manufacturable as light-weight thin plates, but raw material costs for these plates would be prohibitive for transportation and stationary commercial applications. Non-noble, lightweight metals like aluminum and titanium can be used as bipolar plates if the surfaces are protected against corrosion by an electrically conductive, oxidation-resistant barrier coating or cladding. This invention describes compositions and methods of providing such a coating or cladding on a metal like aluminum and titanium.

6. **Briefly describe how this invention solves the problem or meets the need:**

Coating and cladding protective layers on aluminum have been developed which are stable under fuel cell operating conditions. The coating is a graded composition of titanium nitride alloyed with aluminum nitride deposited on a titanium sub-layer on aluminum by magnetron sputtering. The optimum average composition is $Ti_{0.5}Al_{0.5}N$. The addition of Al to Ti reduces the density of d-electron states and therefore the oxidation stability of the coating. The electrical conductivity is also reduced relative to TiN by the addition of Al, but remains very high at the above composition. Typical resistivities are below 1 mohm.cm. Key elements of this invention are: (a) the use of a near-zero bias magnetron sputtering process for deposition in order to obtain coatings with only fine-scale porosity, (b) the use of a chemical anodization procedure involving dipping in sulfuric or chromic acid for a short time followed by boiling in water for 30 minutes to 1 hr to infiltrate the fine scale porosity with internal layers of alumina through the thickness of the coating, and (c) deposit a sub-layer of Ti on the Al substrate to provide a more noble surface on which to coat with TiAlN.

The protective cladding layer material employed is a commercially available exfoliated graphite material known as Grafoil (sold by Union Carbide). The key element of this invention is the process of bonding grafoil on aluminum using a graphite paint (graphite particles in an epoxy resin thinned by an organic solvent, such as toluene) followed by annealing in hydrogen at a low temperature. The high chemical inertness of graphite coupled with its high electrical conductivity

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provides the key properties required for a proton exchange membrane fuel cell bipolar plate.

Potentiodynamic corrosion testing on TiAlN/Ti coated and grafoil cladded plates displayed in Fig. 1 shows corrosion currents corresponding to extremely low corrosion rates of 0.1 and 1mil/year respectively at a potential of 900 mV versus a standard calomel electrode. Long term potentiostatic data for a TiAlN/Ti coated plate in Fig. 2 indicates that a low corrosion current is maintained over a testing period of ten days. Fig. 3 shows polarization voltage versus current density curves for a fuel cell with TiAlN/Ti coated aluminum bipolar plates tested without any significant drop in performance for over 750 hrs. of continuous testing.

7. <u>Describe how to make and use the invention.</u>

TiAlN/Ti coated aluminum plates will be made by first fabricating the metal plates with machined or stamped flow field patterns and gas inlets. The plates will then be degreased, dried and finally reductively plasma-etched in the reactor before deposition on both sides by an unbiased magnetron sputtering process. The as-received coated plates will then be dipped either in conc. sulfuric acid or chromic acid, washed and then boiled in water for 30 minutes prior to use in a fuel cell stack.

Grafoil cladded aluminum plates will be made by applying graphite paint to cleaned aluminum plates with machined gas inlet holes. Grafoil sheets of the same shape and basal dimensions of the aluminum plates will be positioned on the aluminum plates and stamped by a mold with the required flow field pattern and gas inlets – the latter matching those on the plates. The bonding process of grafoil will be repeated for the opposite of each plate. The plates will then be stacked up in a furnance and annealed in 10% hydrogen at 250 C for 1 hr. After cooling in hydrogen, the plates will be ready for use in a fuel cell stack.

8(a) To the best of your recollection <u>what is the earliest date on which you conceived this invention?</u>

[redacted] initiating work on magnetron sputtering of TiAlN/Ti on aluminum, and [redacted] on grafoil cladded aluminum.

(b) <u>Are there any documents or records which confirm this date? If so, please identify:</u>

PEM team weekly report on above dates and notebook A2846.

(c) <u>Are there co-workers or others who can corroborate this date? If so, please identify:</u>

Members of PEM fuel cell team in Torrance and in Morristown – Tim Rehg (Torrance) and MoShu Yang (Morristown)

(a) **To the best of your recollection, what is the earliest date at which the invention was reduced to practice?**

[redacted] and [redacted] for TiAlN/Ti on Al and grafoil on aluminum respectively.

(b) **Are there any documents or records that confirm this date? If so, please identify?**

PEM fuel cell group biweekly reports on above dates and notebook A2846.

(c) **Are there co-workers or others who can corroborate this date? If so, please identify:**

Tim Rehg

10(a) **Did this invention arise in a program that is funded in whole or part by the U.S. government, another company, or any entity other than AlliedSignal?**

_____ YES   x____ NO

(b) **If so, please identify the program and the entity sponsoring the program:**

(c) **Government contract number (if applicable)**

11(a) **To your knowledge is this invention subject to any agreements between AlliedSignal and a third party (e.g. a secrecy agreement, license agreement, joint development agreement, etc.)?**

_____ YES   __x__ NO

(b) **If so, please identify the agreement and the other party:**

12. **You have a duty to disclose to the U.S. Patent and Trademark Office all relevant prior art of which you are aware. Please list all such prior art (e.g. patents, publications, brochures, prior products of ourselves and others) known to you. If a prior art search has been done, it should be included. Briefly indicate how this invention is different from the prior art.**

US Patent 5,624,769 (April 29, 1997) assigned to General Motors Corporation filed on Dec 22, 1995 describes the use of TiN coatings with an intermediate barrier of stainless steel rich in chromium, nickel and molybdenum on Ti or Al for a corrosion resistant PEM fuel cell.

13(a) **Has the product or process which is the subject of this invention been disclosed or communicated to anyone outside of AlliedSignal or the general public?**

_____YES __x__NO

(b) **If so, please provide details including dates and relevant documents:**

(c) **Are you aware of any intent to disclose or communicate the subject of this invention to anyone outside of AlliedSignal or to the general public in the near future? If so, please explain and give the approximate date of disclosure:**

No.

14(a) **Has the product or process which is the subject of this invention been sold or offered for sale to anyone outside of AlliedSignal or to the general public?**

_____YES _____x_NO

(b) **If so, please provide details including dates and relevant documents:**

(c) **Are you aware of any intent to sell or offer for sale the subject of this invention to anyone outside of AlliedSignal or to the general public? If so, please explain and give the approximate date of sale or offer to sell:**

No.

15(a) **Does this invention relate to any other i) issued patents, ii) pending patent applications, or (iii) previously submitted invention disclosures, of AlliedSignal?**

_____YES __x___NO

(b) **If so, please identify. If this is an improvement on an earlier invention, please indicate:**

Send to:

AlliedSignal Law Department
Patent Group
[Relevant Address]
[Phone & Fax]

Signature:

____________________**(name)**
____________________**(phone)**
____________________**(FAX)**

AlliedSignal

Invention Disclosure

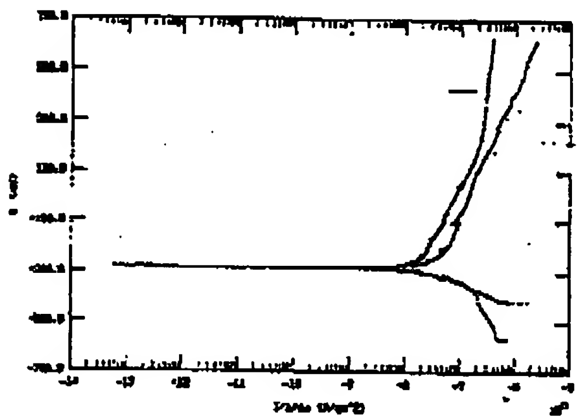


Fig.1: Potentiodynamic Tafel plot of TiAlN/Ti on Al (upper left) and grafoil cladded Al (upper right) using mercury/mercury sulfate reference electrode. 900 mV vs SCE corresponds to 525 mV in this measurement.

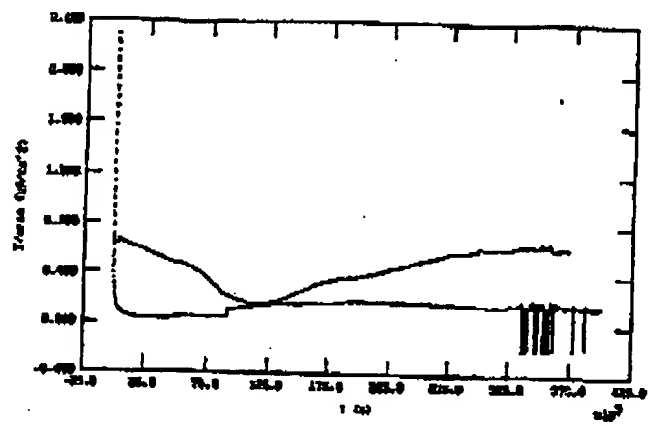


Fig. 2: 10 day potentiostatic corrosion test for magnetron sputtered and chemically anodized TiAlN/Ti on Al at a potential of 900 mV versus SCE using mercury/mercury sulfate as reference electrode. The top curve is for week 1. The bottom curve for week 2 shows an initial decline in corrosion current and a stable value after the first few days.

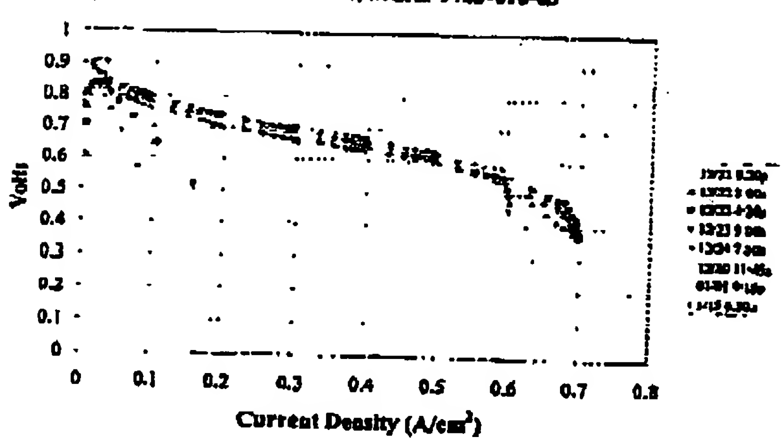


Fig. 3: Polarization data from single cell fuel cell test using TiAlN/Ti coated aluminum bipolar plates

EXHIBIT B [redacted]

PEM Fuel Cell Bipolar Plate Development – Biweekly report [redacted]

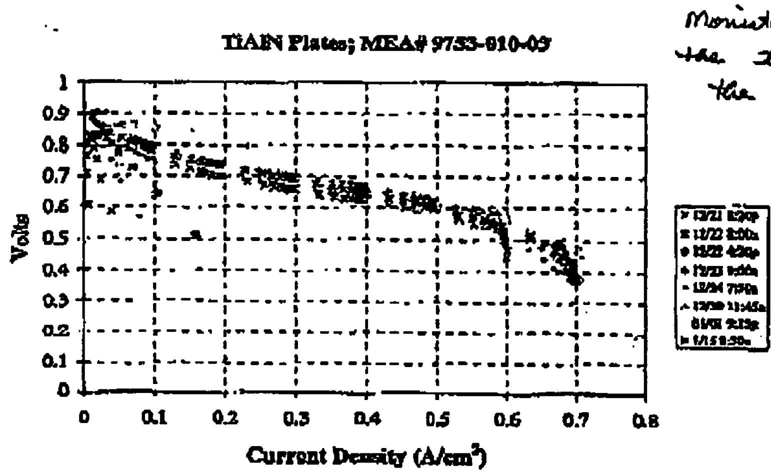
Task 1.2.1.6- Coated Metal Bipolar Plates

1.2.1.6.3 - Determine viability of aluminum as base metal

- We have performed potentiostatic testing at 900 mV versus SCE on a magnetron sputtered TiAlN/Ti coated aluminum plate that was treated by dipping for 1 min. in chromic acid (prepared by dissolving 9% by weight of sodium dichromate in conc. sulfuric acid). The corrosion current decreased to below 10^{-6} amps/cm^2 after the first few days and then remained steady. However, after four days of testing spikes were observed followed by increased corrosion current, indicating that the coating had been suddenly breached. Similar but larger spikes had been previously observed for a plate treated with nitric acid. Because of this result we employed only sulfuric acid treatment for the coated aluminum plates used for the long-term fuel cell test. It was pointed out by Dave Narasimhan that according to some results in the literature, chemical treatment should be followed by boiling in water for 30 min to 1 hr to stabilize the anodization layers formed in the fine scale porosity. It is expected that stabilization would also occur during fuel cell operation at 85 C, but it would be safer to boil the plates in water after the acid treatment to prevent early breaches in the coating. We have prepared samples of TiAlN/Ti coated aluminum that were treated in both sulfuric and chromic acid for 1 minute and then boiled in water for 30 min after washing. These plates will be potentiostatically tested for corrosion.
- Long-term fuel cell testing under low stoichiometry conditions using magnetron sputtered TiAlN/Ti coated aluminum plates have now been under continuous operation for 28 days or 672 hrs without loss in performance. Selected data are shown in Fig. 1. Water at pH 6 has been collected at various times and is being tested for Al and Ti content. We will initiate a performance comparison test with a similar double serpentine flow field graphite bipolar plate under low stoichiometry conditions after completing 1000 hrs of testing.

1.2.1.6.4 - Evaluate alternative approaches

- Chemical anodization of a magnetron sputtered TiAlN/Ti coated 316 stainless steel plate was performed by dipping in chromic acid (prepared by dissolving 9% by weight of sodium dichromate in conc. sulfuric acid) for 1 minute followed by washing and then boiling in water for 30 min. No change in electrical conductivity of the plate was observed. Potentiodynamic corrosion testing will be performed and the result will be compared with that for the as-received coated plate.



Monistown had to shut down the test station to re-fill the humidifiers

Tests

① Pictures before and after to look for pitting

② Analyze the effluent from the fuel cell, ICP analysis

③

Fig. 1: Polarization data from single cell fuel cell test using TiAlN/Ti coated aluminum bipolar plates

- Bipolar plate coupons were fabricated using a modified grafoil/aluminum approach for potentiodynamic and potentiostatic corrosion testing. Grafoil was bonded to aluminum with graphite paint under a small load and the composite was then annealed for 1 hr in 10% hydrogen/argon at 250 C. A good bond to aluminum was obtained with 2-point through-plane resistivity equivalent to pure aluminum. Well-bonded graphite-painted aluminum plates with resistivity lower than that of graphite have been obtained by Jeff Yang from a vendor. These samples have been chemically anodized in chromic acid and boiled in water after washing. Both untreated and anodized samples will be corrosion tested.

2-4 week forward plans:

- Complete 1000 hr fuel cell test using TiAlN/Ti coated aluminum plates, chemical analyses of effluents, and surface and resistivity testing of the plates
- Initiate graphite cell test under similar conditions
- Initiate corrosion testing of grafoil, graphite-painted aluminum and TiAlN/Ti on steel approaches
- Initiate coating of full size Al plates for sub-scale stack testing

EXHIBIT C [redacted]

PEM Fuel Cell Bipolar Plate Development – Biweekly report [redacted]

Task 1.2.1.6- Coated Metal Bipolar Plates

1.2.1.6.3 - Determine viability of aluminum as base metal

- Potentiostatic measurements at 900 mV versus SCE were performed using a mercury sulfate reference electrode at pH 5 for 10 days for as-received and chemically anodized (2 min. concentrated sulfuric acid) plates of Ti-Al-N/Ti coated aluminum prepared by unbiased magnetron sputtering. The potentiostatic data for the as-received sample (Fig. 1) show a number of spikes (probably caused by localized pitting), but on an average the corrosion current remains low. However, in the second week the spikes become numerous and the corrosion current shows a substantial increase. Pitting marks are evident on the surface of the sample. The data for the chemically anodized sample (Fig. 2) shows an immediate drop due to passivation and a stable period of around 4 days followed by a slow increase in corrosion current. In the second week the corrosion current remains stable at an extremely low value after an initial decrease. Chemical analyses of the solutions, SEM examination and quantitative measurements of the electrical conductivity of the samples are in progress.
- We are also pursuing a second chemical anodization route involving dichromate treatment. We have performed 1-2 min. chemical anodization of Ti-Al-N/Ti on Al using a solution of sodium dichromate in nitric acid. We observed no change in the high electrical conductivity of the Ti-Al-N/Ti coating. According to the patent literature this process has provided a high degree of corrosion protection for both aluminum and magnesium. Potentiostatic corrosion testing on this sample has been initiated. The results will be compared with the data discussed above for conc. sulfuric acid treated samples.
- The second single cell stand to be used for testing the coated metal bipolar plates has been put through various calibration runs successfully. Ti-Al-N/Ti coating of the flow field plates are in progress at MultiArc.

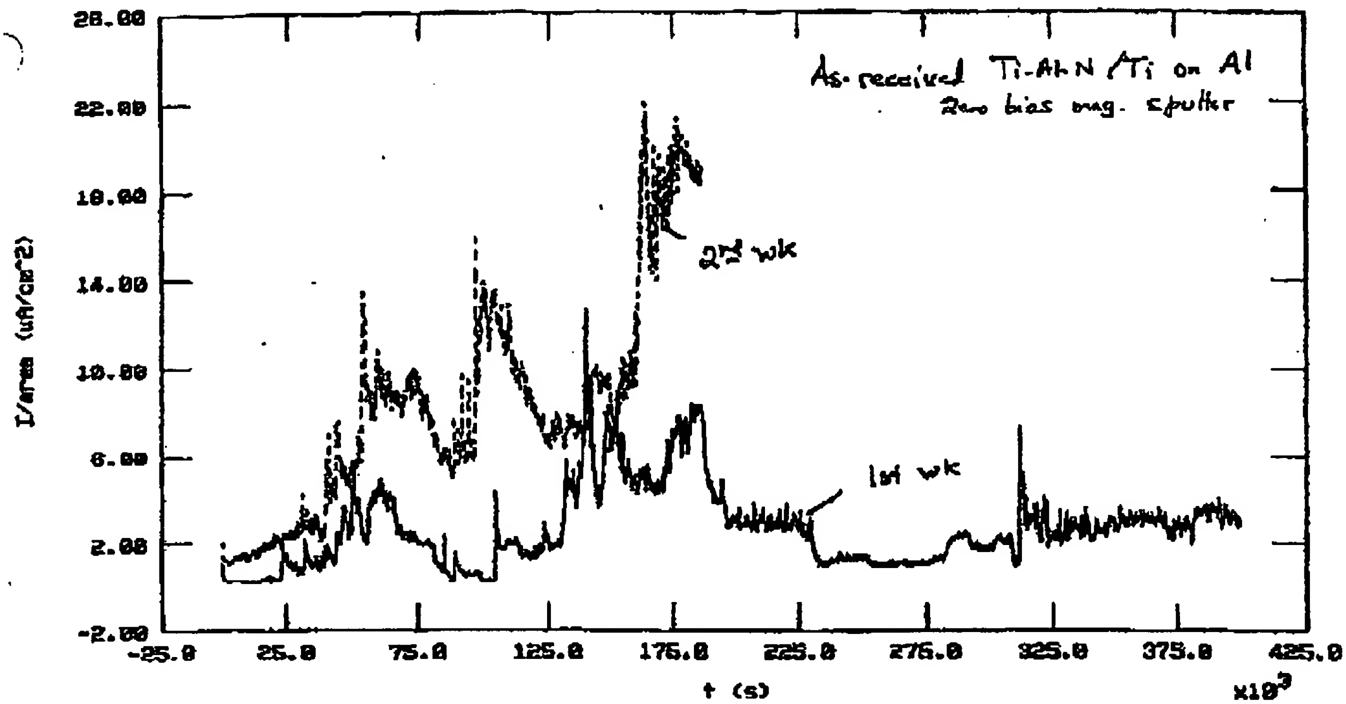
1.2.1.6.4 - Evaluate alternative approaches

- We have thinned a commercial graphite paint (obtained from Acheson Products) with toluene by sonication and used it to paint-coat aluminum and titanium bipolar flow field plates. The coating obtained is optically dense and very conductive. Quantitative measurements of the conductivity and corrosion before and after chemical anodization are in process.
- We also propose to use the modified graphite paint to seal the porosity in grafoil and use this grafoil to laminate aluminum or titanium plates to form grafoil/metal/grafoil

thin composite bipolar plates. Another possible use of the graphite paint would be as a surface coating to enhance the surface conductivity of our conductive plastic bipolar plates.

2-4 week forward plans:

- Complete post-corrosion test chemical, conductivity and microscopic analyses of Ti-Al-N/Ti coated aluminum
- Initiate fuel cell testing of Ti-Al-N/Ti coated aluminum plates
- Complete corrosion testing of dichromate treated Ti-Al-N/Ti on aluminum
- Complete potentiodynamic corrosion testing of graphite painted aluminum and Ti-Al-N/Ti on 316 stainless steel
- Seal grafoil with graphite paint and perform permeation tests



Potentiostatic testing with Hg/Hg₂SO₄

900 mV vs SCE or
520 mV vs Hg/Hg₂SO₄

Fig. 1

10 day continuous testing.

Spiking is occurring. More spikes during second week. Pitting is occurring, visible to the eye. These data do not have H_2SO_4 treatment.

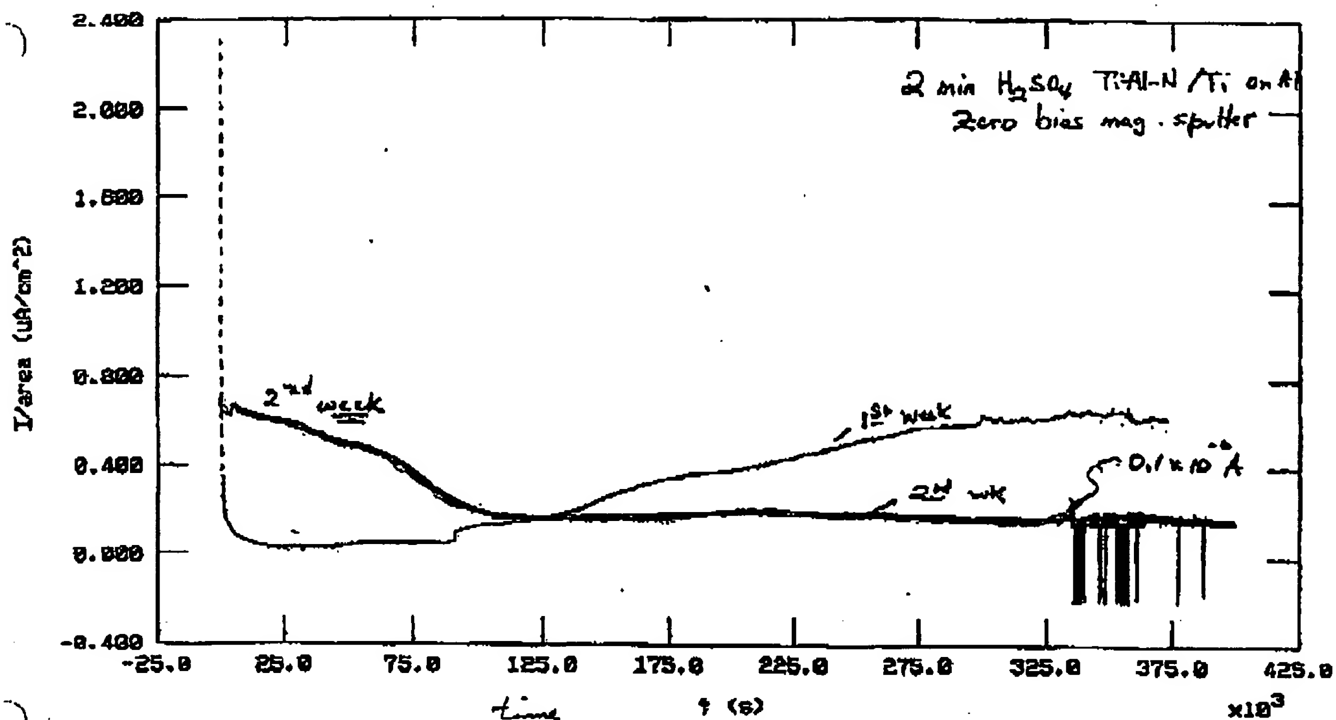


Fig.2

These data reflect a treatment with H_2SO_4